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Spectral Hole Burning Memory Using InAs Self-assembled Quantum Dots

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This paper describes observations of spectral hole burning of InAs self-assembled quantum dots (QDs) embedded in a pin diode. At 5 K, a narrow spectral hole of photocurrent corresponding to the reading light laser with a width of less than 1 nm was observed. At a writing light power of 8 mW, the spectral hole depth increased as the electric field was increased. The spectral hole was observed up to 40 K. The spectral hole width at 8 mW fitted well with the convolution integral of a Gaussian distribution for the reading light and a Lorentzian distribution for the absorption change taking into account a homogeneous broadening of InAs QDs of ≤ 80 µeV. The spectral hole lifetime at 8 mW was estimated to be in the order of 10^{-6} s from a rate equation. Type II QD structures were proposed to prolong the hole lifetime. The optical absorption spectrum of a 15-stack InAs QD structure was also observed at 77 K and 300 K.

1. Introduction

The quantum dot (QD) has attracted much interest in both low-dimensional physics[1] and applications in electronic and optical devices.[2,3] Fabrication of self-assembled QDs using the Stranski-Krastanow growth mode[4] has advantages compared to other methods[5,6] in that it is maskless, almost dislocation-free, and uses high-density QD formation techniques. Self-assembled QDs are also applicable to optical devices that operate from the visible to near infrared range with various materials.[7,8] Usually, mainly due to the size fluctuation along the growth direction, self-assembled QDs show a photoluminescence (PL) line that is as wide as 80 to 100 meV.[9] This large inhomogeneous broadening (Γ_i) is suitable for a wavelength-domain multiplication optical memory[10] or hole burning memory. This is because QDs can be considered to have narrow homogeneous broadening (Γ_h) due to a δ-function-like density of states which gives a large wavelength-domain multiplicity of Γ_i/Γ_h. However, the hole burning of self-assembled QDs, which is a nonlinear optical effect in the 0-dimension, has not been studied well.

In this paper, we report on observations of spectral hole burning in the photocurrent spectra of InAs self-assembled QDs[11] and discuss the characteristics of the spectral hole. We also describe further approaches to improve the characteristics of these QDs.

2. Experiments

Figure 1 shows the structure of the sample used in this experiment. Five stacked InAs QDs layers were inserted into a pin GaAs diode with 20 nm GaAs interval layers to isolate the QD layers electronically. Figure 1 also shows the corresponding band diagram at thermal equilibrium. Each QD layer is formed as a nominal 1.8 monolayer of InAs and has a dot density of 5×10^{10} cm^{-2}. The wafer was mesa etched to a diameter of 500 µm and was fabricated with a non-alloy ohmic contact on top of a heavily p-doped layer. Another contact to the n-type region was fabricated at the

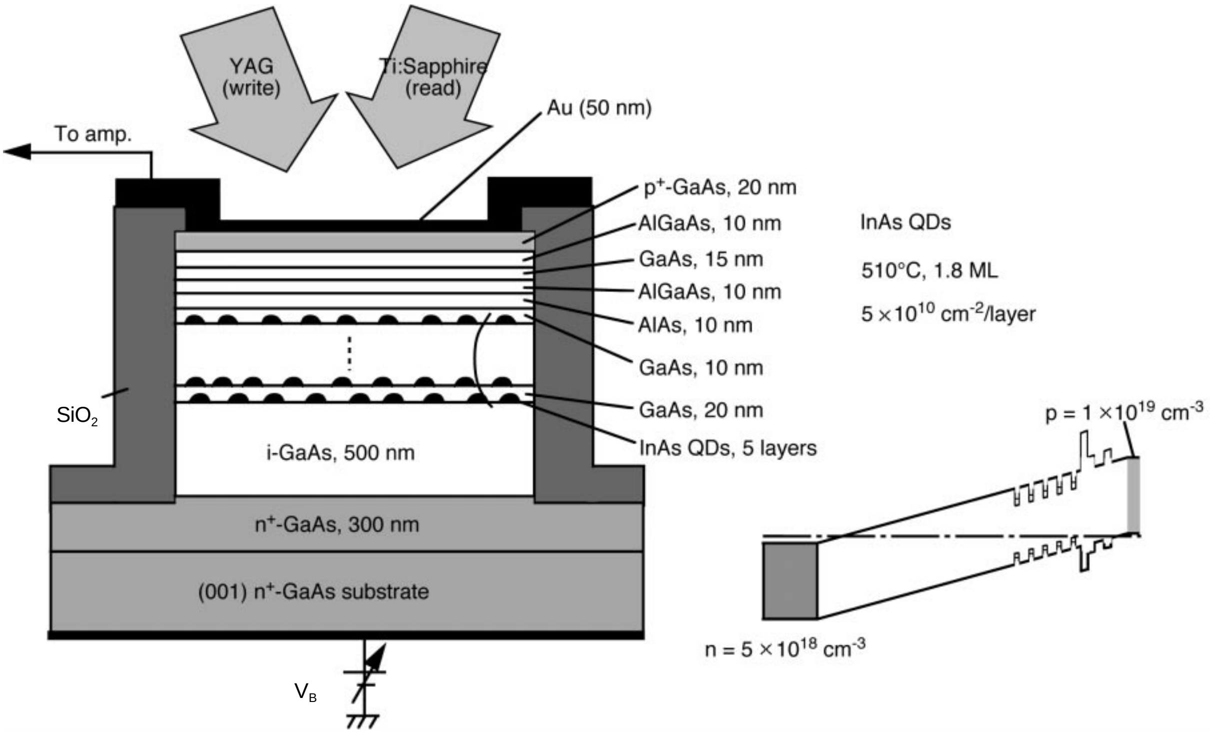


Figure 1
Structure of sample used in experiments. Five layers of InAs self-assembled quantum dots (QDs) were embedded in pin diode. Diameter of sample is 500 µm.

bottom of the substrate. To increase the lifetime of the spectral hole, an electric field was applied to enable either electron or hole tunneling from the QDs. The top contact on the p-GaAs was made transparent with a thin (50 nm) Au film to enable irradiation of the QDs. As a reference, a pin diode with no InAs QDs was also fabricated.

The writing light source was a continuous-wave (CW) 1,064 nm YAG laser and the reading light source was a tunable CW-Ti:sapphire (TiS) laser. The bandwidth of the TiS laser was ≤ 40 GHz (165 µeV). The bandwidth of the YAG laser was ≤ 6.4 GHz (27 µeV) and contained several lines. The writing laser power was varied from 0 to 20 mW, and the reading laser power was ≤ 80 µW. Both lasers were directed unfocused onto the sample. The photocurrent corresponding to the tunable TiS laser was detected with a lock-in amplifier through a current amplifier to eliminate the photocurrent due to the CW YAG laser. During the photocurrent measurements, the wavelength of the TiS laser was monitored using a wavemeter.

3. Hole burning spectroscopy of InAs QDs

Prior to the hole burning measurements, the PL and photocurrent spectra of the sample were measured. **Figure 2** shows the photocurrent spectrum, which is equivalent to the absorption spectrum, obtained at 300 K with a WI_2 lamp. The broad absorption bands related to the InAs QDs were well represented by three Gaussian curves. For example, the ground state fitted with a full width at half maximum (FWHM) of 82.5 meV. The higher two peaks are those of the GaAs and wetting layer and fit with Lorentzian curves. The peak PL energy at 300 K was 40 meV below that of the ground state absorption. This energy difference is caused by carrier redistribution due to thermal energy.12) The PL spectrum at 77 K can be considered to correspond to the ground states

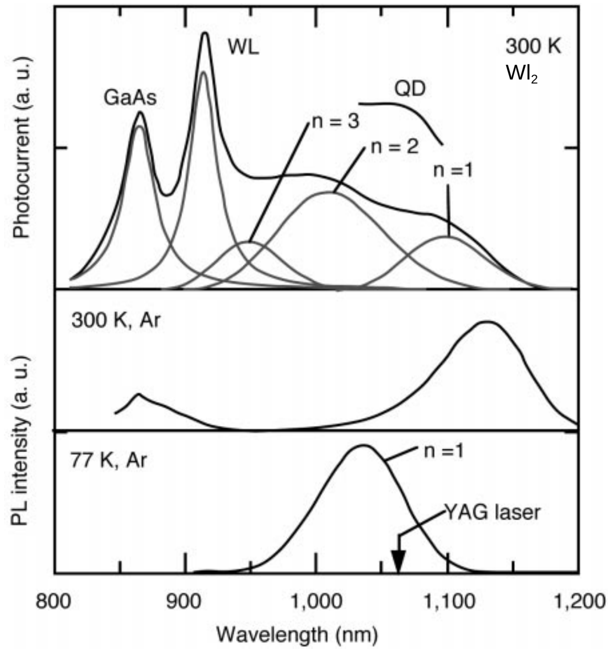


Figure 2
Top) Photocurrent spectrum measured with WI_2 lamp at 300 K with fitted Gaussians for QDs and fitted Lorentzian for wetting layer and GaAs.
Middle and bottom) Photoluminescence (PL) spectra at 300 K and 77 K with Ar laser excitation.

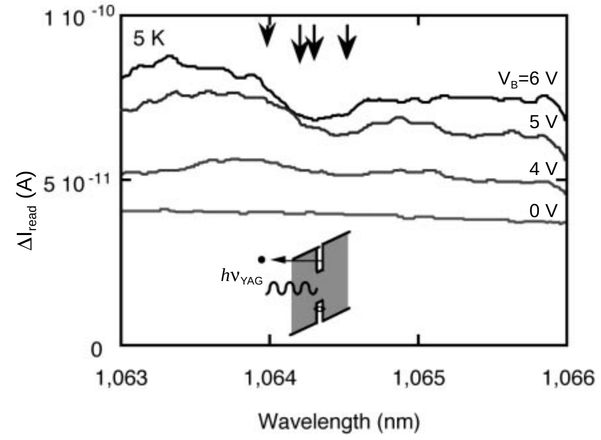


Figure 3
Dependence of photocurrent spectra, I_{read}, on bias voltage (V_B) at 5K for 8 mW irradiation from CW YAG writing light laser. Reading light source was CW Ti:Sapphire (TiS) laser. Arrows indicate the wavelengths of YAG laser.

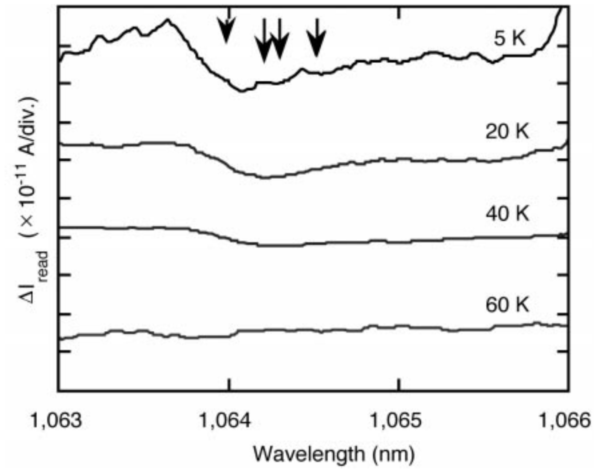


Figure 4
Difference between I_{read} with and without irradiation by YAG laser at 5 K, 20 K, 40 K, and 60 K. YAG laser irradiation power was 8 mW. Arrows indicate wavelengths of YAG laser.

of the QDs because the sample was excited under a weak condition and thermal carrier distribution to excited states was suppressed compared to the case at 300 K. The ground state absorption obtained by fitting to the photocurrent spectrum at 300 K agreed well with the 77 K PL spectra after the slight blue shift due to the temperature change of band gap. Therefore, the photon energy of the YAG laser was estimated to be lower than the peak energy of the ground state distribution.

The photocurrent response, I_{read}, to the TiS lasers was measured under a YAG laser irradiation of 8 mW and a TiS lasers irradiation of 32 µW. **Figure 3** shows the external bias voltage (V_B) dependence of I_{read} at 5 K. As V_B increases, a dip appeared at the wavelengths of the YAG laser lines indicated by the arrows. The dip deepened as V_B was set to 4 V and higher.

This result can be interpreted as follows. Increasing V_B increases the internal electric field, which stimulates a larger number of photogenerated carriers in the QDs to tunnel from the ground state through the triangular potential barrier formed by the field. Electrons are more likely to tunnel because of their smaller effective mass. Here, the carriers are electrons, and heavy holes can be considered to stay in the QDs, as shown in the inset of Figure 3. The residual holes inhibit additional optical absorption of the TiS laser light under intense irradiation of the YAG laser. As the electron tunneling rate becomes comparable

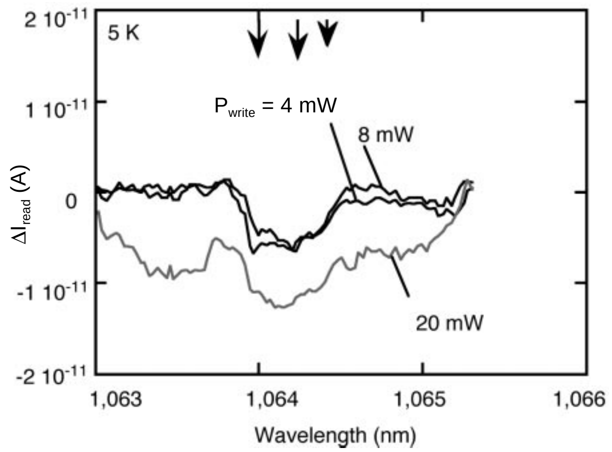


Figure 5
Dependence of I_{read} on writing power, P_{write}, at 5 K as extracted from I_{read} without irradiation by YAG laser. Arrows indicate wavelengths of YAG laser.

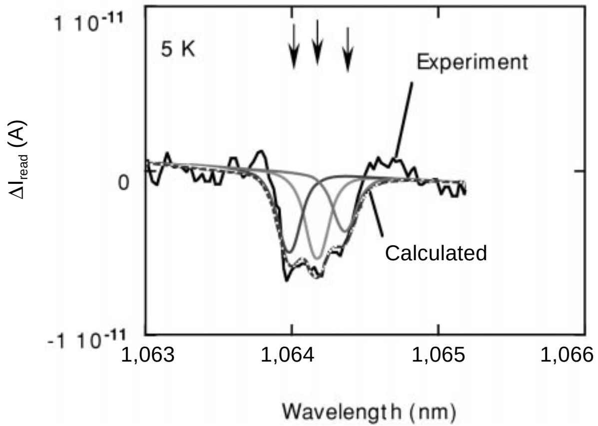


Figure 6
Comparison of spectrum observed in experiment at 5 K with writing YAG laser power of 8 mW and spectrum calculated from convolution integral with Gaussian for reading TiS laser and Lorentzian for absorption in QDs.

with or larger than the radiative recombination rate in the QDs, we can expect to see a spectral hole appear in I_{read}. Such behavior was not observed with the reference sample. Therefore, the dip represents the formation of a spectral hole which originates from the InAs QDs.

Figure 4 shows the temperature dependence of the difference, ΔI_{read}, between I_{read} at a YAG laser irradiation of 8 mW and I_{read} with no YAG irradiation over the range from 5 K to 60 K at a bias voltage of 6 V. As the temperature increased, the hole depth decreased and eventually vanished at 60 K. This suggests that carriers were thermally excited, possibly to the wetting layers. A similar excitation of carriers has been reported in QD lasers.13) However, the onset of thermally assisted tunneling cannot be excluded. Further study is needed to find a way to suppress the phonon interaction and improve the operation temperature of self-assembled QDs.

Figure 5 shows the writing power dependence of ΔI_{read} at 5 K. The difference in the emission lines of the YAG laser was from the use of different YAG lasers. The spectral hole is similar at 4 mW and 8 mW, but is broader at 20 mW. It is possible that at 20 mW, the increased irradiation led to an increased number of acoustic phonons and that broadening of the spectral hole was due to light absorption by these phonons.

4. Discussion of spectral hole

4.1 Spectral hole width

To investigate the spectral hole width, we performed numerical fitting to the experimental spectra. We took into account the bandwidth of the writing and reading lasers. The YAG lasers used in this experiment have several emission lines that are 0.1 to 0.2 nm apart and less than 6.4 GHz (27 μeV) wide. These emission lines are caused by multiple transition levels in the YAG crystal or longitudinal mode limited by cavity length. On the other hand, the CW TiS reading laser has more closely spaced emission lines which can be approximated by a single Gaussian distribution with an estimated width of ≤ 40 GHz (165 μeV). We applied a Gaussian function for the CW TiS laser spectrum and a Lorentzian function for the absorption spectrum of the QDs for each emission line of the YAG laser and then calculated the convolution integral between these two types of functions. In the calculation, we used an FWHM of 165 μeV for the Gaussian function, and the FWHM of the Lorentzian function, Γ, was used as a fitting parameter. **Figure 6** shows the calculated and experimental results for a writing power of 8 mW at 5 K. Taking into account the three emission lines of the writing YAG laser that were measured by a high-resolution monochromator, the calculated curve fitted well with the experimental result, which cannot be fitted with any single Gaussian or Lorentzian function. The obtained Γ was about 100 μeV. Assuming a Lorentzian distribution for both the YAG laser spectra and the absorption spectrum of the QDs, Γ_h was estimated to be ≤ 80 μeV. This value may be too high because the experimental hole width is usually $2\Gamma_h$ for Lorentzian absorption spectra.[14)]

The experimental curve in Figure 6 indicates the presence of "anti-holes" on both sides of the spectral hole; these anti-holes are typical signs of photophysical hole burning. They can also be seen in Figures 3, 4, and 5. The formation mechanism and relationship of anti-holes with the spectral hole in energy is not yet clear. However, we must study and understand the origin of anti-holes before we can apply self-assembled QDs to hole burning memories.

4.2 Spectral hole lifetime

The lifetime of the spectral hole can be derived from the time-evolution of the hole depth. However, the writing and reading lasers constantly irradiated the sample and it takes much longer to measure the corresponding I_{read} than the spectral hole lifetime. This makes it difficult to directly determine the lifetime from the hole spectrum. Therefore, we used a rate equation on a carrier in a steady state.

We assumed that resonantly photogenerated electrons escape much faster than holes from the QDs, so that only holes could be treated with the rate equation, and that two holes can occupy the ground state in a QD due to the spin degrees of freedom. In a steady state, the equation can be written as follows:

$$\frac{\sigma n}{\tau} = c\frac{P}{S}\left(1-\frac{\sigma}{2}\right)n \qquad (1),$$

where σ is the probability of hole occupancy at the ground state of the QD, τ is the recombination time of holes and equals the spectral hole lifetime, n is the number of QDs irradiated by the YAG laser, P is the incident power of the laser, S is the incident area of the laser, and c is a coefficient proportional to P having the dimensions of cm^2/Ws. The left term shows the recombination rate and the right term shows the generation rate. The photocurrent, I_{ph}, can be also written as follows:

$$I_{ph}(P) = qcP\,(1-\sigma/2)\,n \qquad (2)$$

$$= \frac{2qnS}{\tau}\left(1-\frac{1}{1+cP\tau/2S}\right) \qquad (3),$$

where Eq. (3) is derived from Eq. (1). The incident power of the reading laser is two orders of magnitude smaller than that of the writing laser. The I_{read} which is not subtracted by I_{read} without YAG laser irradiation can be written as follows:

$$I_{read}(P) = \left.\frac{\partial I_{ph}(P)}{\partial P}\right|_{P=P_{write}} \cdot P_{read} \qquad (4)$$

$$= \frac{qcn}{\left(1+\dfrac{cP_{write}\tau}{2S}\right)^2} P_{read} \qquad (5),$$

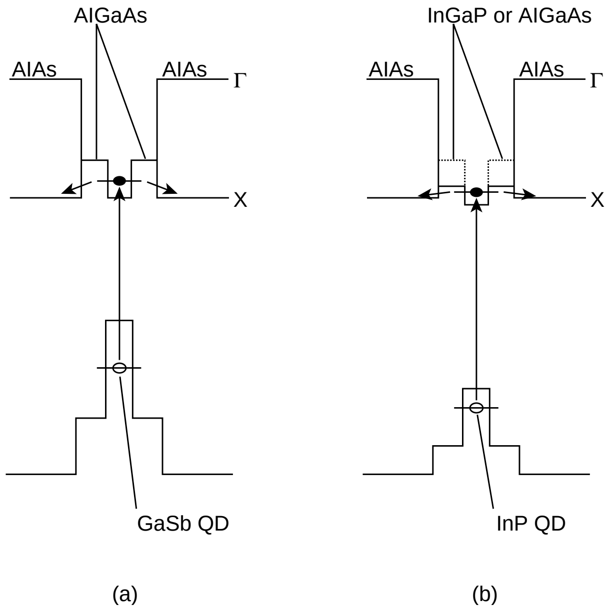
where P_{write} is the incident power of the writing laser and P_{read} is that of the reading laser. c can be derived from Eq. (5) using the experimental result of $I_{read}(P_{write}=0)=25$ pA. Since the laser beam was not focused on the sample surface, P_{read} of 32 μW was calibrated by the squared ratio of the sample diameter (500 μm$^{\varnothing}$) to the beam diameter (≈ 2 mm$^{\varnothing}$). The obtained c was 2.3×10^5. The experimental parameters of $n=5 \times 10^{10} \times 5 \times$ (27 μeV/82.5 meV), $I_{read}(P_{write}=8$ mW$)=19$ pA, and S ≈ 2 mm$^{\varnothing}$, yeilded a τ of 1.2×10^{-6} s.

5. Approaches to improve hole burning memory using self-assembled QDs

5.1 Type II quantum dots

For the practical use of self-assembled QDs in a hole burning memory, the hole lifetime must be increased. However, in QDs that use a type I heterostructure such as InAs/(Al)GaAs, increasing the reverse bias that is applied to the pin diode to extract electrons from the QDs generates an undesirable increase in dark current. This causes some of the information to be lost due to recombination of residual holes with electrons and also reduces the hole lifetime.

To solve this problem, we propose a self-assembled QD having a type II heterostructure. This structure does not need an external bias perpendicular to the hetero-interface to extract electrons from the QDs and may exhibit a drastically reduced dark current. **Figure 7** shows the energy band diagrams for a GaSb QD and InP QD which can be grown by MBE, GSMBE, or MOVPE. Photogenerated electrons tunnel to the X-point in the AlAs through the potential barrier and are spatially separated from the holes, which reduces the recombination rate. Even a small bias that is parallel to the hetero-interface may contribute to a further decrease in the recombination rate. In such a case, GaAs could be used instead of AlAs and the spectral hole lifetime could be increased. Using this structure, hole burning by all-optical measurement can be also achieved.



(a) (b)

Figure 7
Schematic band diagrams of QD with type II heterostructure for (a) GaSb/ AlGaAs/AlAs system and (b) InP/ InGaP(or AlGaAs)/AlAs system. Dotted line in conduction band in (b) indicates case for AlGaAs tunneling barrier.

5.2 Multiple stacked QD structures for all-optical measurement

If the spectral hole lifetime is much longer than the estimated lifetime, it becomes difficult to measure the spectral hole directly from the photocurrent, and all-optical measurement will be required. A single QD layer is less suitable for measurement of the spectral hole due to the small SN ratio. The SN ratio of a hole burning memory using QDs increases with the number of QDs[10]; however, the surface coverage of QDs per layer is as low as 15% for an InAs QD of 5×10^{10}cm^{-2}. Therefore, a multiple-stacked QD structure is needed to obtain a satisfactory SN ratio. Although an InAs/GaAs QD is not always appropriate for observing a persistent spectral hole burning phenomena (see Section 4.2), using this material, it is easy to build a multiple-stacked QD structure and examine the possibility of all-optical measurement of stacked QD layers. There are only a few reports on the optical absorption spectrum of self-assembled QDs, and further study of hole burning memories is required.

Figure 8 shows the optical absorption spectra of a multiple-stacked InAs/GaAs QD structure with 15 QD layers at 77 K and 300 K. The interval layer thickness is 20 nm, and TEM observation does not show a vertical alignment of QDs.

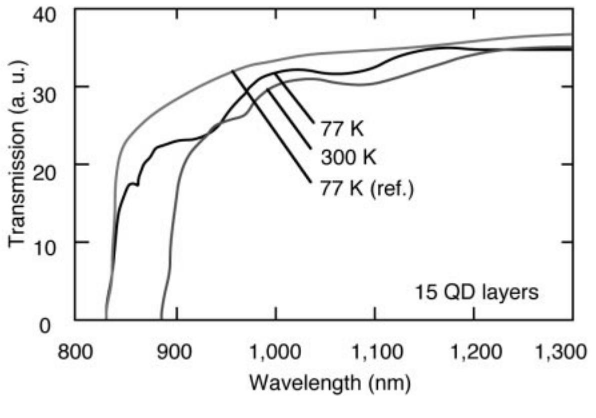


Figure 8
Optical absorption spectra of multiple-stacked InAs QD structure with 15 QD layers at 77 K and 300 K. Dotted line shows spectrum of n-GaAs substrate at 77 K as reference.

After observing the spectrum of the n-GaAs substrate, the broad peaks at 930 nm and 1,080 nm at 77 K appear to originate from the InAs/GaAs QDs. The peak energy of the first excited state is more influenced by temperature than that of ground state; this may be due to the temperature-dependence of strain in the InAs/GaAs QD system and the difference in the degree of confinement. The reason for the difference in the positions of the QD peaks in Figures 2 and 8 is not yet clear. The shoulder at 860 nm corresponds to the absorption of the wetting layer, which peak is also observed in a single QD layer by PL. A similar spectral structure was also observed with 10 QD layers. This result shows the possibility of direct measurement of the spectral hole by all-optical measurement using a multiple-stacked QD structure.

5.3 Design estimation for hole burning memory

We examined the design for a self-assembled QD hole burning memory using the model proposed by Muto.10) We treat optical method for writing and reading processes. We used a type II QD structure as an absorption media. The transmitted light through the QD media was detected by a photodetector which generated a photocurrent. The signal was the difference in the photocurrents generated by photon flux through the marked and virgin areas. The noise is the root mean square of the shot noise fluctuations. Details are given in Reference 10).

If we assume that the S/N is greater than 10, the bleaching of QDs in the writing process is perfect, and the photon flux for reading does not cause substantial power broadening of the detected spectral hole, then the peak absorption cross section, $\sigma_{s,}$ of the QDs is as follows:

$$\sigma_S < 1.88 \times 10^{-5} Af(\alpha_0 L)^2 \qquad (6),$$

where $$f(\alpha_0 L)=\frac{1-\exp(-\alpha_0 L)}{\sqrt{1+\exp(-\alpha_0 L)}} \qquad (7)$$

and A is the area of the reading laser beam, α_0 is the initial absorption coefficient, and L is the sample layer thickness. The multiplexing factor (MPF), defined as N_{tot}/N_ω (where N_{tot} is the total QD density and N_ω is the QD density within the homogeneous broadening at $\hbar\omega$), with these parameters using $\alpha_0 = \sigma_s N_\omega$ is given by:

$$MPF / L = N_{tot}\, \sigma_S / \alpha_0 L \qquad (8).$$

An N_{tot} of 3.5×10^{16} cm^{-3} is obtained for an average QD density per layer of 7×10^{10} cm^{-2} and an interval layer thickness of 20 nm. If we assume that $\sigma_s\Gamma_h = 10^{-16}$ 10) and A=2 μm$^{\o}$, then MPF/L is 17.3, 12.3, and 5.6 for Γ_h =100 μeV, 200 μeV, and 1 meV, respectively. If we set the MPF to 50 (20) for each Γ_h, L values of 2.9 (1.06), 4 (1.6), and 8 (3.58) μm are needed for QD layers of 145 (58), 203 (81), and 446 (177), respectively.

After accounting for the growth procedure and reproducibility, 100 QD layers is a realistic target. Γ_h shows a temperature dependence of approximately $\exp^{T/31}$,15) meaning that hole burning memory operation at 77 K would be unlikely at present. However, a hole burning memory with a spectral hole width of ≤ 200 μeV at ≤ 40 K would be feasible. In this temperature range, several 10s or 100s of G-bit hole burning memory could be possible. The temperature dependence of Γ_h may be due to acoustic photon interaction with the QDs, which might be inherently unavoidable. If this is the case, a major innovation will be needed to solve this problem.

The hole burning memory is a type of ROM, but it requires a refresh procedure to assure memory retention. Assuming an average access time of 50 nm per bit, a complete sequential refresh for a 100-Gbit memory would take 5,000 seconds. Therefore, in a 100-Gbit memory, each element including the frequency domain would need to be able to maintain its condition for this period. If we use the type II QDs mentioned in Section 5.1 at 40 K, stability for 5,000 seconds would require a separation energy of 95 meV between the electron ground state in the QDs and the X-point of the AlAs.[10] The Γ-point of GaSb is about 0.1 eV above that of GaAs,[16] which is comparable with the X-point of AlAs. A GaSb/AlGaAs QD structure can be considered to have a higher degree of potential confinement in the conduction band than an InAs/GaAs QD due to its large Al content of about 0.5. This implies that the GaSb/AlGaAs/AlAs QD system could satisfy the stability requirement and therefore could be used as a practical hole burning memory.

6. Summary

We investigated spectral hole burning in the photocurrent spectra of InAs self-assembled quantum dots embedded in a pin diode. The spectral hole appeared at bias voltages above 4 V and at temperatures below 40 K. The spectral hole has a writing power dependence which shows saturation broadening at 20 mW. The spectral hole can be regarded as being the combination of several holes which correspond to the YAG laser emission lines. The width of homogeneous broadening obtained from numerical fitting was less than 80 μeV. The hole lifetime was estimated to be in the order of 10^{-6} s. The use of self-assembled QDs having a type II heterostructure using GaSb or InP in a hole burning memory was proposed to increase the hole lifetime. The optical absorption spectra of a multiple-stacked InAs QD structure were clearly observed at 77 K and 300 K, suggesting the possibility of all-optical measurement of a hole burning memory.

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